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A SIMPLE APPARATUS FOR THE DETERMINATION OF EFFECTIVE RADIUM CONTENT OF RADIUM-BEARING SUBSTANCES

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A SIMPLE APPARATUS FOR THE DETERMINATION OF EFFECTIVE RADIUM CONTENT OF RADIUM-BEARING SUBSTANCES

by

M.G. Grenier*

ABSTRACT

A simple device was designed to determine the radium content of materials such as backfill from uranium mill tailings. The apparatus operates on alpha-counting principles and consists of a sample holder permanently coupled to a scintillation cell. The device is simple, inexpensive and easily built. This method is attractive where a large number of samples have to be processed routinely. Effective radium contents can be calculated three days after the sample is placed inside the apparatus. The error margin for measurements is less than 10%.

Key words: Radium analysis; Radon emanation; Backfill; Uranium mines.

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MISE AU POINT D'UN APPAREIL D'UNE GRANDE SIMPLICITÉ POUR DÉTERMINER LE CONTENU EFFECTIF DE RADIUM DE MATIÈRES CONTENANT DU RADIUM

par

M.G. Grenier*

RÉSUMÉ

On a mis au point un dispositif d'une grande simplicité pour déterminer le contenu de radium de matières telles la terre de remblayage provenant des installations de traitement de résidus d'uranium. L'appareil fonctionne d'après le principe de comptage des particules alpha et comprend un support d'échantillon raccordé de façon permanente à une cellule scintillante. Le dispositif est simple, peu coûteux et facile à construire. Cette méthode peut être avantageuse quand un grand nombre d'échantillons doit être traité régulièrement. Le contenu effectif de radium peut être calculé trois jours après que l'échantillon a été placé dans l'appareil. La marge d'erreur dont il faut tenir compte quant aux mesures est inférieure à 10 % .

Mots clé: Analyse du radium; émanation de radon; terre de remblayage; mines d'uranium.

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INTRODUCTION

The effective radium-226 content of soil or backfill material is a useful quantity, especially when evaluating the impact of storing such materials on the immediate environment. This quantity is the activity of radium, in a given sample, releasing radon-222 to the atmosphere upon decay.

Measurement of the radium content of soil and water samples is not new, and several techniques have been developed to that end (1,2). Barretto et al. (1) used the sealed-can gross alpha-counting approach. In this method, the sample is sealed inside an ingrowth flask of known volume and is allowed to emanate radon for a period of 8 to 10 days. This flask is then connected to an evacuated alpha-scintillation cell for radon concentration measurements. This method is accurate, but requires careful handling and timing.

The sealed-can gamma-spectroscopy method used by Austin (3) is very similar, but is an improvement over the alpha-counting method since the need to transfer a radon gas sample from the emanation cell is eliminated. Gamma-spectroscopy apparatus is, however, very sophisticated and comparatively expensive.

This work describes a simple, inexpensive apparatus for the determination of radium content in materials. This apparatus was found to be effective when assessing effective radium content of moist materials, such as backfill from uranium tailings used to fill mined-out areas in underground mines. The method combines the simplicity of the alpha-counting methods, without the need for transferring a gas sample as in the gamma-counting method. The accuracy associated with the instrument is approximately 10%, and comparison with traditional emanation methods have shown that the method is in agreement within less than 5%. On the other hand, the method is much simpler by comparison and has definite advantages where several samples have to be

analyzed on a routine basis.

DESCRIPTION OF APPARATUS AND OPERATION

The apparatus which consists of two chambers linked by openings is shown in Figure 1. The approximate overall dimensions are 17 cm in height and 10 cm in diameter. The material used was 0.5 cm thick Plexiglas. The bottom portion of the cell is the scintillation chamber. It is 7.6 cm in diameter and the inside walls are coated with scintillation material (ZnS, silver activated). The bottom disc is kept clear and is the surface in contact with the photomultiplier tube when the cell is placed in the alpha-counting instrument (Figure 2).

The top portion of the cell contains the sample to be analyzed. Radon produced in this volume diffuses to the bottom section via the two openings described above. The backfill sample is placed in a perforated sample tray. The perforations are small enough to hold sample grains within and they allow radon to emanate from the sample in all directions. The sample tray is placed on a perforated sample platform held in place with O-rings which allow the tray to be moved up or down to accommodate various sizes of samples. A threaded Plexiglas cover sealed with an O-ring encloses the top portion after the sample is inserted. A Swagelok type connector was fitted onto the cover to allow the volume of the cell to be deduced using an apparatus described elsewhere (4).

To determine the effective radium content of a sample, a known mass of backfill material is placed in the sample tray, the cover is screwed shut and the time is noted. In a static configuration such as this one, it is a simple task to note the growth of alpha-activity. Using this data and the theory described in the next section the effective radium content may be calculated.

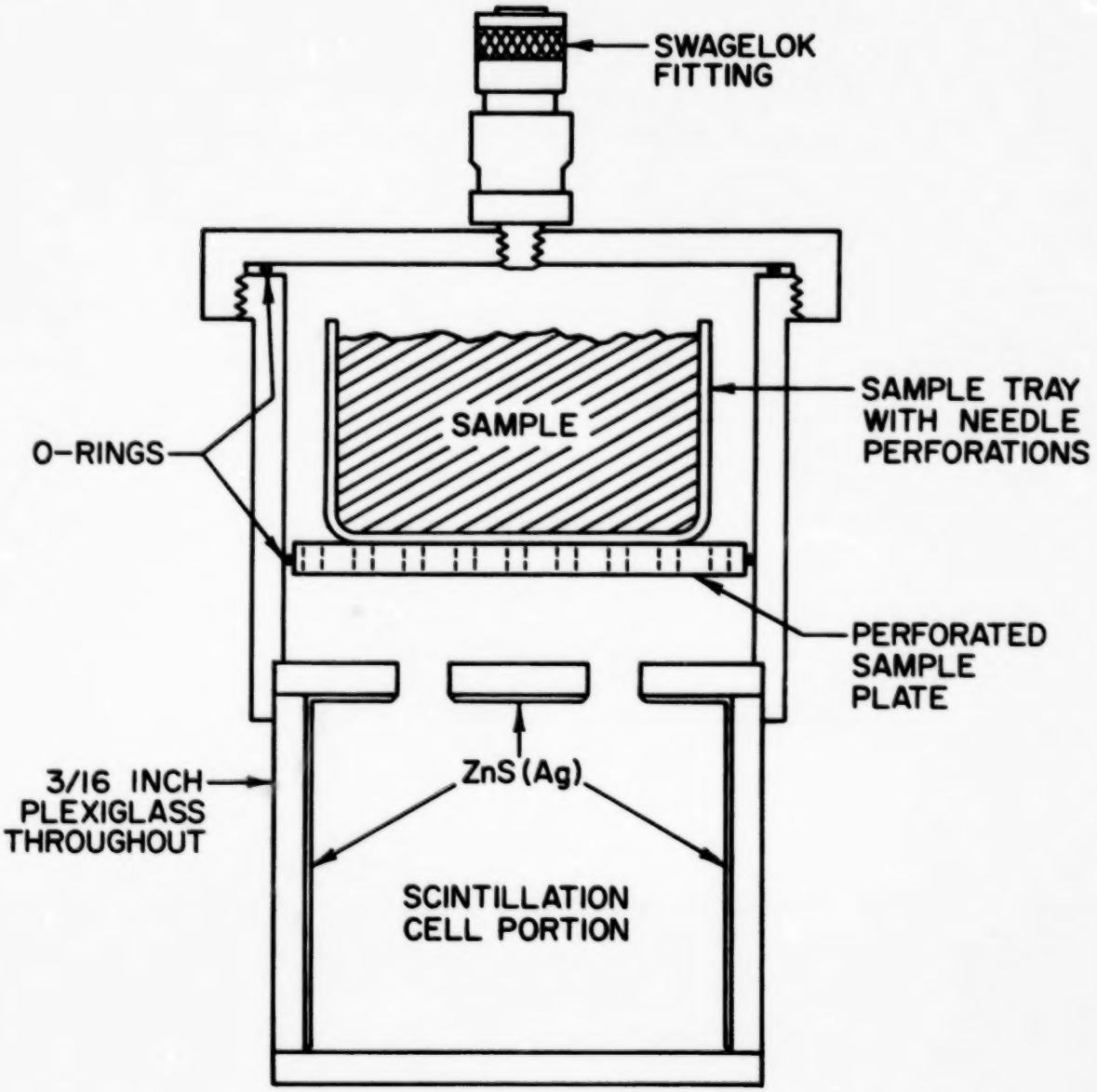

Fig. 1 - Apparatus designed to measure the effective radium content of samples. The cell is constructed of .5 cm thick Plexiglas, is 17 cm in height, and approximately 10 cm diameter.

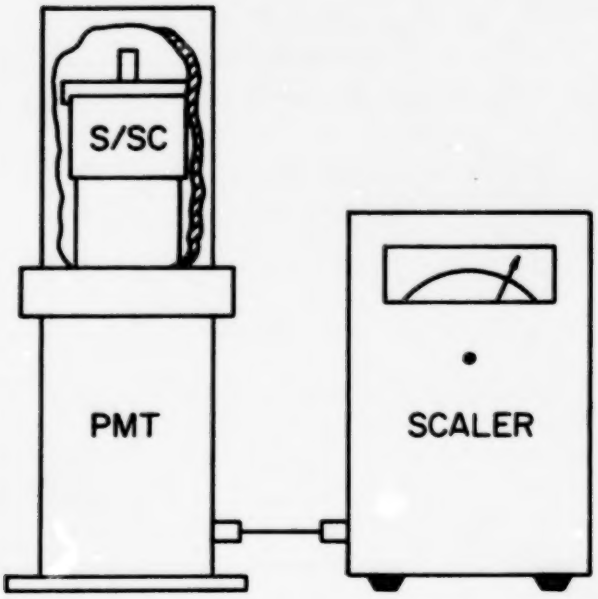

Fig. 2 - Diagram showing the emanation cell (S/SC) along with other required apparatus. The cell is enclosed in a photomultiplier assembly (PMT) connected to a scaler.

THEORY

When a radium bearing substance is placed in a volume that contains no radon gas initially, the growth of the gas is expressed by:

$$I_{Rn} = I'_{Ra} (1 - \exp(-\lambda_{Rn} t)) \qquad \text{Eq 1}$$

where, I_{Rn} is the activity of radon-222 (Bq)

I'_{Ra} is the effective radium-226 activity of the sample which contributes to the release of radon-222 (Bq)

λ_{Rn} is the decay constant of radon-222 (0.000126 min^{-1})

t is the time in minutes from the beginning of the experiment.

Using Equation 1 and assuming equilibrium between radon-222 and its short-lived decay products, the effective radium concentration of a sample (Bq/g) may be found after an ingrowth period of three to four days, using Equation 2:

$$I'_{Ra} = \frac{cpm(t)\ V_2}{180\ \varepsilon\ V_1\ m\ (1 - \exp(-\lambda_{Rn} t))} \qquad \text{Eq 2}$$

where, cpm(t) is the alpha-count rate at time t, from the decay of the radon and daughter mixture at equilibrium

V_1 is the volume of the scintillation portion of the apparatus (0.173 L)

V_2 is the total apparatus volume (0.39 L)

ε is the efficiency of counting of the cell in conjunction with the scaler being used (0.66)

m is the sample mass (77.9 g).

PERFORMANCE

A 77.9 g sample of uranium mine backfill was placed and sealed inside the apparatus. The cell was then placed inside the photomultiplier

tube/scaler assembly where the growth in activity was monitored for a period of more than 500 hours (20.8 days). These data are shown graphically in Figure 3. Also shown in this graph is the theoretical growth calculated using Equation 1.

Both curves agree well. Furthermore, Equation 2 can be used to establish the effective radium content as early as three to four days after the beginning of the experiment. Theoretically, it should be possible to perform the calculation sooner, but it was necessary to wait a few days in order to obtain a larger count rate. This reduced counting statistic errors and by that time, the assumption made above concerning radon and daughter equilibrium is a reasonable one.

Using the values given with Equation 2 for the various physical parameters, the effective radium content of a 77.9 g sample of backfill was calculated as 0.82 Bq/g. Knowing the accuracy of measuring the volumes of the cell sections and taking counting statistics into account, an error of 10% is associated with the effective activity quoted above.

DISCUSSION

The advantages of the system described here are that only one photomultiplier tube/scaler unit is required. The only limitation on the number of daily samples that can be processed is the number of cells available. There is also no need to transfer radon to a separate scintillation cell for counting since the scintillation portion is inherently part of the apparatus. Less handling is required and the method is simplified. Cells may be sealed and left alone for a few days at which time they can be counted for alpha activity. This method was found to be very convenient when a large number of samples needed to be analysed on a routine basis.

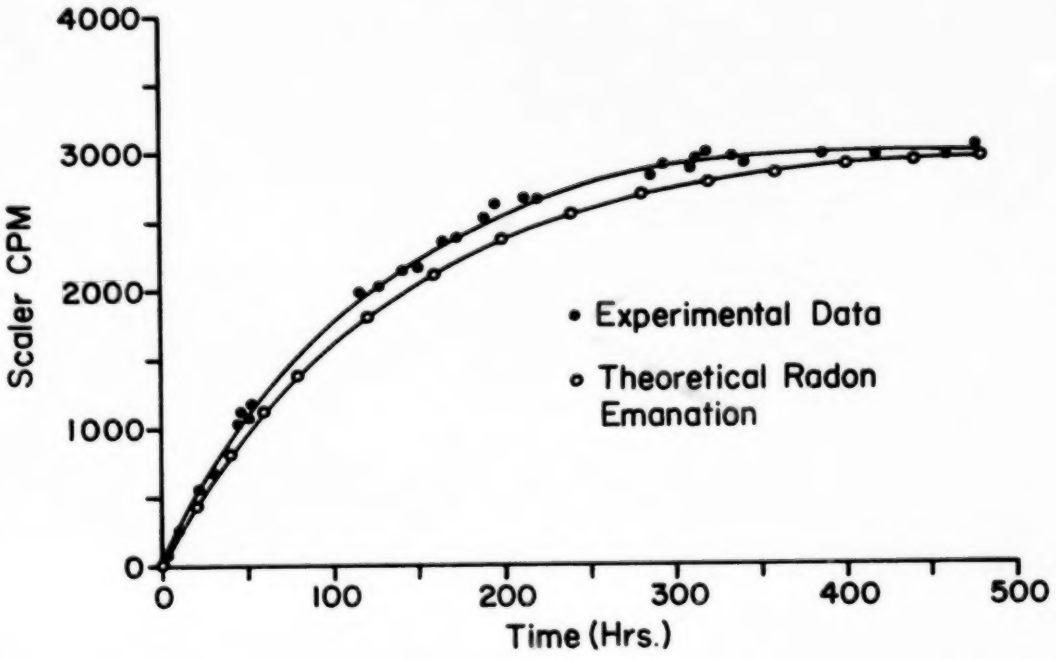

Fig. 3 - Graph showing the theoretical radon growth curve with the cell. Also shown is the actual growth curve.

The system, however, has a 10% range of accuracy and care must be exercised when moving the cell. Sample material spilled from the tray may contaminate scintillator material in the bottom portion of the cell.

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